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IONIZATION IN LAMINAR FLAMES

- USSR -

by N. N. Inozentsev

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FOREWORD

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IONIZATION IN LAMINAR FLAMES

[Following is a translation of an article by N. N. Inozemtsev in the Russian-language periodical Izvestiya Akademii Nauk SSSR -- Energetika i Avtomatika (Bulletin of the Academy of Sciences USSR -- Power Engineering and Automation), No. 2, Moscow, 1960, pages 59-66.]

At present it is possible to consider the presence of unbalanced ionization in the front of a laminar flame as proved. In addition, a number of works have suggested that the formation of ions in a flame is connected with the nature of the chemical process of combustion [1-3]. In this connection, obtaining experimental confirmation of this proposition would make it possible to judge the velocity of a chemical reaction and the dimensions of a zone of intense chemical change in laminar and turbulent flames by the value and the width of the bump in the ionization current. It is obvious that in order to prove the proportionality of the ionization current to the velocity of the chemical reaction it will be necessary to make precise quantitative measurements of the unbalanced ionization current in laminar flames and to determine the most important characteristics of the ionization process (concentration of ions and electrons, the rate of formation of electrons, the coefficient of recombination, and others), depending upon the initial parameters of the burning mixture. However, very material difficulties are encountered in measuring the current of ionization in flames on account of the small width of the zone of intense ionization and the high temperature in the flame front. Moreover, the absolute value of the current of ionization is very small, which causes additional difficulties in amplifying and recording this value.

The methods used for investigating ionization in flames are quite varied, but not all of them are suitable for quantitative evaluation of ionization in the laminar front of a flame. Thus, the method of using Langmuir probes employed in cited works [2, 3] is applicable only under high-vacuum conditions since the theory of a probe presupposes the lack of collisions between charged particles and molecules reaching the probe through a space charge layer. Moreover, this method is applicable for comparatively great concentrations of charged particles ($>10^8$ per cubic centimeter), otherwise the probe will not be screened from the surrounding discharge by a layer of the space charge [4]. Langmuir probes are suitable when the potentials on them are close to the potential of the charged space.

The ionization in the front of a laminar flame cannot be measured by the attenuation of radio waves on account of the small size of the front, which is only a fraction of the length of the electromagnetic waves used for this purpose (~1 centimeter). This method is also not too sensitive in case of weak ionization when the ion concentration is less than 10^{11} per cubic centimeter, therefore it is used primarily for measuring ionization of the combustion products of flames with an admixture of alkali metals [2, 5].

Determining ionization by the deviation of the lines of the current under the action of an electric field gives rise to doubt due to the great complexity of calculations since this method presupposes the determination of the value of the product of the ion concentration in the reaction zone and the width of this zone by the measured curvature of the line of the current in the electric field and, moreover, the effect of a strong electric field which curves the entire cone of the flame may be material.

Determining the ionization by measuring the conductivity of the flame between two electrodes requires knowledge of the mobility of the electrons.

At present it is well known that the negatively charged particles in a flame of gas-air mixtures are free electrons [1, 2-5]. The values of the mobility of electrons under different conditions can be calculated theoretically with sufficient accuracy [4].

All the methods enumerated above for investigating ionization yield different values of concentrations of charged particles in the front of hydrocarbon flames (from 10^6 to 10^{12} per cubic centimeter). This is explained apparently by the inapplicability of some methods under flame conditions and the inaccuracy of other methods due to errors in the pick-ups and recording apparatus used.

This article contains a method for investigating ionization in flames with two-contact pick-ups, the results obtained from using this method to study hydrocarbon-air Bunsen flames under different initial conditions, and an analysis of the results obtained.

The Method of Measurement. The ionization current in the front of the flame was measured by two-contact pick-ups supplied by direct current batteries. The pick-ups were two-channel ceramic pick-ups with steel wire contacts inside arranged in such a way that only their end surfaces would come in contact with ionized particles. The external diameter of the ceramic insulators varied from 1 to 3 millimeters while the diameter of the contacts varied from 0.2 to 0.5 millimeters. The power supply battery set up a constant electric field between the contacts of the pick-up, and ions were collected from the ionized zone of the flame under the influence of this field. The pulse of ionization current I_i obtained when the front of the flame was intersected by this pick-up set up an input voltage at the ends of the scaled resistance $U_{in} = I_i R_s$ (Figure 1a) which was amplified first by a preliminary one-tube alternating current amplifier (the pulse was fed into the grid of a 6N3P tube) then fed into the input of the amplifier of a 25I cathode pulse oscillograph.

The output voltage was fixed by a camera from the oscillograph screen. The electron tube of the standard 25I oscillograph was replaced by a 13-L037 tube with prolonged yellowish-green afterglow which ensured the reliable fixation of the ionization process on the photographic film at the high frequencies prevailing in rapid movements of a thin laminar front and in turbulent flames (moreover, special, highly sensitive film was used).

Since special attention was devoted to precise quantitative evaluation of ionization, all possibilities of this apparatus were checked.

Frequency and amplitude characteristics were taken, the effect of the value of the scaled resistance R_s was tested, and the errors caused by the conductivity of the ceramic insulation of the pick-up were also analyzed. The frequency and amplitude characteristics of the apparatus are shown in Figures 1b and 1c; the frequency characteristic has a horizontal section in the frequency range of 30-40 cycles to 10^6 cycles per second. This passband makes it possible to obtain undistorted reproduction of current bumps when the laminar front is intersected at different rates. The linear part of the amplitude characteristic is the working section in which the input voltage and the ionization current are determined

$$h_{out} \rightarrow U_{in}, \quad I_i = \frac{U_{in}}{R_s}$$

Matching the working section with the linear part of the characteristics was accomplished by the proper selection of the value of the scaled resistance R_s (in our case $R_s \leq \text{megohms}$).

Since any insulating material has some conductivity, errors are possible due to the conductivity of the ceramic insulation of the pick-up, and these errors will increase with an increase in the temperature of the ceramics.

The layout used here excluded the possibility of such errors since an alternating current amplifier was used to amplify the signal from the pick-up, which excluded the direct component of the current and amplified only the alternating signal set up when the pick-up intersected the front of the flame. The conductivity of the ceramic insulation of the pick-up could affect changes in the voltage on the grid of the 6N3P tube, which would cause shifting the working point from the linear part of the characteristic of the tube. When it is cold ($T \approx 20$ degrees Centigrade) the ceramic has a resistance of approximately megohms, which is entirely sufficient for the normal operation of the apparatus. When the ceramic is heated to 200-300 degrees Centigrade, its resistance drops down to approximately 200 megohms. In this case a large role is played by the value of the scaled resistance R_s which, together with the resistance of the ceramic itself, forms a voltage divider for the power supply battery. With a high-resistance input, when $R_s > 50$ megohms, the errors

due to shifting of the working point when the ceramic insulation of the pick-up is heated can become noticeable. A high-resistance input ($R_g = 49.2$ megohms) was used in measuring the thermal ionization of the combustion products. Flames were studied by rapid movement of the pick-up, that is, the pick-up was moved manually with a speed of 1-2 meters per second or mechanically (pneumatic insertion) with a speed of 10-20 meters per second so that the ceramic insulation would not be heated up while being passed through the flame.

A developed picture of the ionization process in the flame front on the given scale and irrespective of the speed of passing the pick-up through the flame, that is, independent of the time, was obtained by the use of a linear semiconductor slide-wire rheostat instead of displacement potentiometer. Thus, this layout makes use of scanning in accordance with the coordinates of the pick-up (the pick-up is connected with the slide-wire of the rheostat). Adjusting resistances included in the circuit of the slide-wire rheostat permit changing the width of the section of the path of the pick-up spread over the entire oscillograph screen, that is, it is possible to fix the ionization in any section of the flame on any scale by the coordinates. If one knows the linear size of the working section of the slide-wire rheostat L_r shown over the entire oscillograph screen, it is easy to determine the value of the ionization current and the size of the ionized zone in the flame by the height and width of the bump on the photographic film.

The Results of Measurements. Current-voltage characteristics were taken in laminar flames, both in the front itself and in the combustion products (Figure 2). Up to the power supply voltage $U_g = 20$ volts, the ionization depends linearly on the voltage on the electrodes of the pick-up, which indicates the equality of the rate of occurrence of electrons in the flame q with the rate of recombination $q = \beta n_+^2$ (the diffusion of electrons is neglected and n_+ is set equal to n_+), and β -- is the coefficient of recombination.

Saturation begins to take place at $U_g = 35$ volts, that is, all the electrons which originate in the flame are carried away by the field of the pick-up, then

$$q = \frac{I_g}{eV}$$

where I_g is the saturation current, e -- the charge on the electron, V is the space from which electrons are collected.

A certain increase in the current when $U_g > 35$ volts is explained by an increase in the space $\sqrt{\text{volume of } V}$ due to a growth in the intensity of the field of the pick-up E .

The concentration of electrons n was determined in accordance with the values of the current in the linear part of the current-voltage characteristic

$$I_1 = en_3 K_3 E F_{el}$$

(where $K_3 \gg K_+$, K is the mobility of the ions), and F_{el} is the area of the electrodes. The rate of emergence of electrons and the coefficient of recombination are in accordance with the value of the saturation current. The average time of recombination is

$$\tau_p = \frac{n_3}{q} = \frac{1}{\beta n_3}$$

If we know the average time of recombination, the mobility of the electrons, and the intensity of the field of the pick-up, it is possible to determine the average path of the electrons along the electric field of the pick-up λ_p where recombination of the electrons does not occur while they are passing along this path

$$\lambda_p = \tau_p K_3 E$$

Then the working space from which the electrons are collected is equal to

$$V = \frac{1}{4} \pi (d_3 + 2\lambda_p)^2 \lambda_p$$

(d_3 is the diameter of the electrodes of the pick-up), that is, the working space is proportional to the intensity of the field, which explains the increase in the current in the current-voltage characteristic in the region of saturation with an increase in U_3 .

The ionization current and the width of the zone of unbalanced ionization in the linear part of the current-voltage characteristic and under saturation conditions were measured in the laminar front of the flame and in the combustion products of propane-air and gasoline-air mixtures. Typical oscillograms recording the ionization current and the position of intersection of the Bunsen flame have been presented in Figure 3 [See Note]. The results of investigation of ionization in the front, depending upon the initial parameters of the mixture, are presented in Figure 4. The concentration of electrons in the front of a laminar flame for the hydrocarbon fuels investigated amounted to $\sim 2.2 \times 10^7$ per cubic centimeter for $\alpha \approx 0.9$, $P = 1.0$ atmospheres, and $T_1 \approx 20$ degrees Centigrade. (Note: A description of the experimental apparatus in which the ionization of laminar flames was investigated is given in the work [6].)

The dependence of the concentration of electrons upon the pressure can be expressed by the exponential law of proportionality

$$n_3 \sim p^{1.82}$$

When calculating the concentration in accordance with measurements of the current, the mobilities of the electrons K_e were determined with Bradbury's formula [4], taking into account losses of energy on the part of the electrons in collisions with molecules during their passage through the electric field of the pick-up.

The rate of production electrons in the flame under normal conditions and $\alpha = 0.9$ was $q = 17 \times 10^{17}$ per cubic centimeter per second and its dependence on the pressure $q \sim P^{1.93}$. The coefficient of recombination $\beta = 3.3 \times 10^3$ cubic centimeters per second and $\beta \sim P^{-1.7}$.

The average time for recombination was found from the calculations to be on the order of $\sim 10^{-10}$ seconds, then $\lambda_p \sim 10^{-3}$ centimeters, which amounts to a tenth of the width of the bump of the ionization current in the laminar front (Figure 4b). Thus, one can speak of the local nature of the study of ionization conducted by this method since the pick-up collects electrons from a narrow zone limited by the linear dimension λ_p , which is of an order equal to the value of the free path of an electron. Therefore, when determining the intensity of the electric field of the pick-up one can make use of the expression for the maximum intensity

$$E_{\max} = \frac{U}{H}$$

(H is the distance between the centers of the electrodes) and the voltage drop with withdrawal from the dipoles for the distance λ_p is not taken into account.

One can neglect errors due to distortion of the electric field of the pick-up by the space charge under the conditions prevailing in the flame since the concentration of electrons is small here: 10^7 - 10^8 per cubic centimeter. In this case one may consider that the charges move independently of each other in the gas in the flame and that the charge per unit volume is so small that the field at any point in the interval between electrodes is determined by the electric field of the electrodes; that is, it is possible to neglect the distribution of charges in the space affected by the electric field of the pick-up. The density of the current under which the field is distorted by the space charge can be determined by comparing the number of lines of force in the field set up by the electrodes with the number of lines from the charges [7].

The charge per unit surface of the flat electrodes is

$$\sigma = \frac{E}{4\pi} = \frac{1}{4\pi} = 80 \cdot 10^{-3} \left[\frac{e\lambda \cdot \text{CGSE}}{\text{cm}^2} \right] \quad (1a)$$

(with a field intensity of the pick-up $E = 1 \left[\frac{e\lambda \cdot \text{CGSE}}{\text{cm}} \right]$).

The charge in the space [volume] corresponding to a unit of the surface of the flat electrodes is

$$en_3H = \frac{HI_1}{K_2E} \quad (1b)$$

Distortion of the field by the space charge should be taken into consideration in those cases when both values of the charges become comparable.

In our case the space charge is equal to

$$\sigma_{os} = en_3H = 4.8 \cdot 10^{-10} \cdot 0.04 \cdot 10^7 \cdot 10^8 = 0.2-2.0 \cdot 10^{-3} \left[\frac{e \cdot CGSE}{cm^2} \right] \quad (1c)$$

that is, the fraction accounted for by space charges under the conditions prevailing in a flame amounts to only 0.25-2.5 percent and the effect of the space charges on the electric field of the pick-up can be neglected.

The concentration of electrons in the combustion products immediately behind the flame front is, according to our measurements, approximately 0.47×10^5 per cubic centimeter under normal conditions -- this is roughly 1/500 the value in the flame front. The rate of production of electrons in the combustion products is $\sim 0.7 \times 10^{14}$ cubic centimeters per second and the coefficient of recombination $\beta \approx 3.0 \times 10^4$ cubic centimeters per second. If we calculate the effective ionization potential by SAGA's equation [4-7] for thermal ionization in accordance with the known degree of ionization in the combustion products $x = n_3/N$ (where N is Loschmidt's number), then we shall obtain $U_1 = 13.4$ ev, that is, the effective potential of ionization is equal to the ionization potentials of the combustion products (CO_2 , H_2O). When vapors of the salts of the alkali metals (for example, sodium) are added to the combustion products, the ionization increases.

Thus, our experiments confirmed the presence of unbalanced ionization in the laminar flame front. The bumps in the ionization current (Figure 3a) remind one of the distribution of the unbalanced concentration of CH , CC , and others in the flame front. The width of the zone of unbalanced ionization δ_1 is, according to our measurements approximately 0.2 millimeters under normal conditions and when $\alpha \approx 0.9$ (Figures 4b and 4e) and depends on the initial parameters of the mixture. When the laminar front of the flame is intersected quickly (with a speed of ~ 10 meters per second) some expansion of the ionization zone is observed, which can be explained by the effect of the dimensions of the pick-up. However, the height of the bump remains constant when the speed of intersection is changed, which confirms the localized nature of this method (the action of the field of the pick-

is spread over a distance roughly equal to the free path of an electron) and the presence of a wide frequency passband for the apparatus, which makes it proper to use this method for studying turbulent flames.

It can be shown on the basis of the relationships obtained that the rate of production of electrons in the front of the flame is proportional to the velocity of the chemical reaction.

For this purpose we shall make use of the derivation of the formula of the normal velocity of the propagation of a flame U_n (obtained by A. G. Prudnikov) for the case of heat release as a function of distance in the front of a flame

$$U_n = \frac{1}{c_p \rho_1 (T_2 - T_1)} \int_{-\infty}^{+\infty} \Phi(x) dx \approx \frac{\sqrt{2\pi} \Phi_{\max}}{c_p \rho_1 (T_2 - T_1)} \delta_1 \quad (1)$$

where Φ_{\max} is the maximum release of heat, $\omega(c_1 T)$ is the velocity of the chemical reaction $\Phi_{\max} \sim \omega(c_1 T)_{\max}$, δ_1 is the width of the laminar front of the flame, ρ_1 is the initial density of the mixture, T_1 and T_2 are the initial and the final temperatures, and c_p is the heat capacity.

Knowing the experimental dependence of the normal velocity on the initial parameters of the mixture [6] and substituting the values found for the rate of production of electrons in the flame q_{\max} and the width of the zone of unbalanced ionization δ_1 for Φ_{\max} and δ_1 , one can calculate the relationship of the experimental value of U_n and the theoretical k

$$k = \frac{U_n c_p \rho_1 (T_2 - T_1)}{\sqrt{2\pi} q_{\max} \delta_1} \quad (2)$$

The results obtained from calculating k for different initial parameters of the mixture are presented in Figure 5. The constancy of the values of k with changes in the composition of the mixture, the pressure, and the initial temperature indicate the presence of a proportionality between the release of heat (or the velocity of the reaction) and the rate of production of electrons in the front of the flame [See Note] and since $q \sim I$, the ionization current measured by the given method is proportional to the velocity of the chemical reaction in the flame. (Note: The different absolute values of k are explained by the fact that quantities which are functions of the given initial parameter of the mixture were taken into account in calculating the relationship of the experimental value of U_n with the theoretical value.)

In addition, experiments were conducted to measure the parameters of ionization and normal velocity when the fresh propane-air mixture was

diluted with carbon dioxide [See Note] (Figure 4c). The equality of the relationship with k for different percentages of CO_2 content in the mixture (by weight) also confirm the proportionality of $I_1 \sim \omega(c, T)$. (Note: The temperature of combustion of the propane-air flame was measured by an infrared pyrometer and the reversal of the spectral lines of sodium.)

Conclusions. Measurements of conductivity with two-contact pick-ups produced quantitative data on the different parameters of ionization in laminar hydrocarbon-air flames and their dependence upon the initial parameters of the mixture. The concentration of ions and electrons in the front of the flame was two orders higher than the thermal concentration in the combustion products. The conclusion of the proportionality of the measured ionization current and the velocity of the chemical reaction was based on an analysis of the dependence of the parameters of ionization in the flame front and the normal velocity of propagation of the flame upon the initial parameters of the mixture.

Submitted 29 September 1959

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FIGURE APPENDIX

6N3P 13-4037

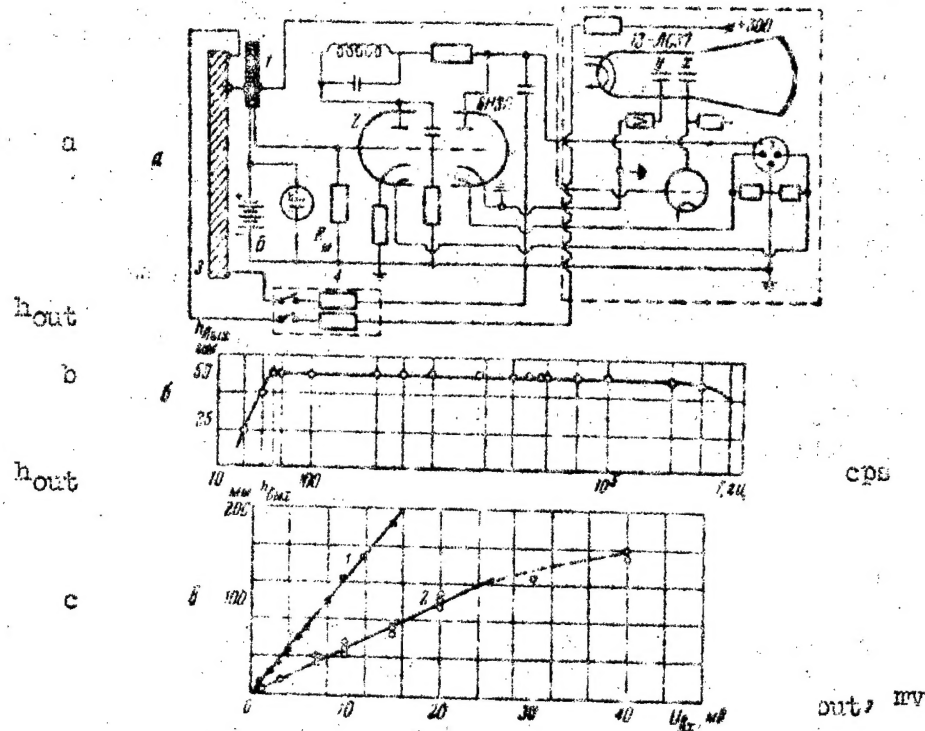


Figure 1. a -- fundamental diagram for measuring ionization in a flame: 1 -- two-contact pick-up, 2 -- one-tube amplifier, 3 -- semiconductor slide-wire rheostat, 4 -- regulating resistances, R_M -- scaled resistance, B -- power-supply battery for the pick-up; b -- diagram of frequency characteristic; c -- diagram of the amplitude characteristic; 1 -- input resistance of the oscillograph 1:1; 2 -- input resistance of the oscillograph 75 ohms.

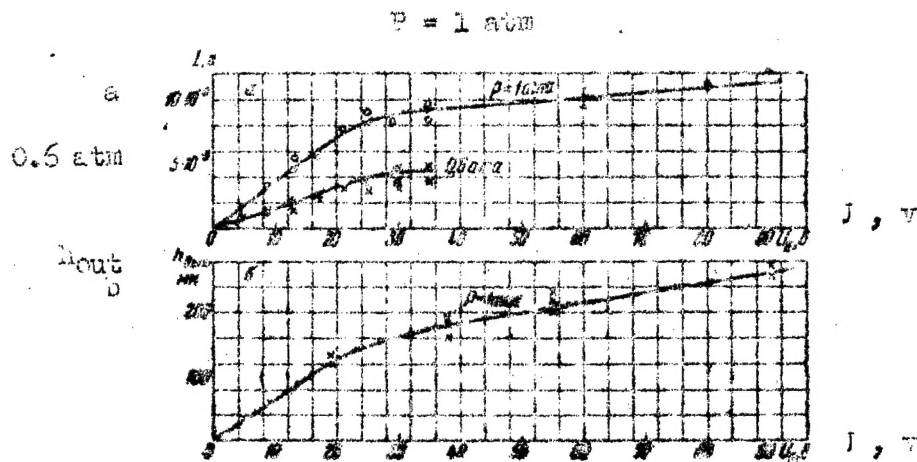


Figure 2. Current-voltage characteristics of propane-air flame:
 a -- in the front of the flame (pick-up with a diameter of 1 millimeter, $T_1 = 20$ degrees, $\alpha = 0.9$); b -- in combustion products (pick-up with diameter of 3 millimeters; $T_1 = 20$ degrees C, $\alpha = 0.9$)

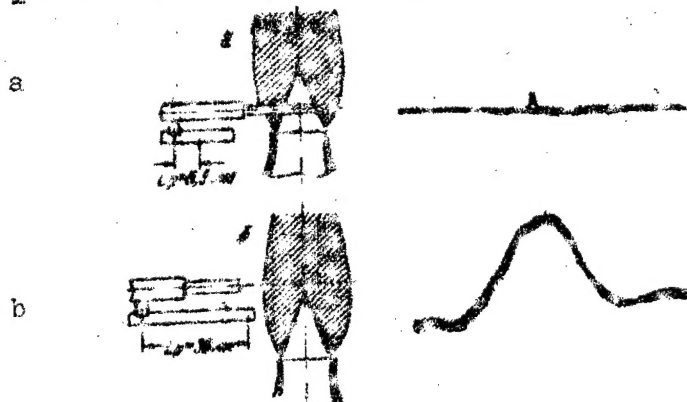


Figure 3. Oscillograms of the ionization current in a laminar flame:
 a -- in the front of the flame (pick-up with a diameter of 1 millimeter, $U_g = 4.5$ volts, $R_M = 1.2$ megohms), b -- in the combustion products (pick-up with a diameter of 3 millimeters, $U_g = 70$ volts, $R_M = 49.2$ megohms)

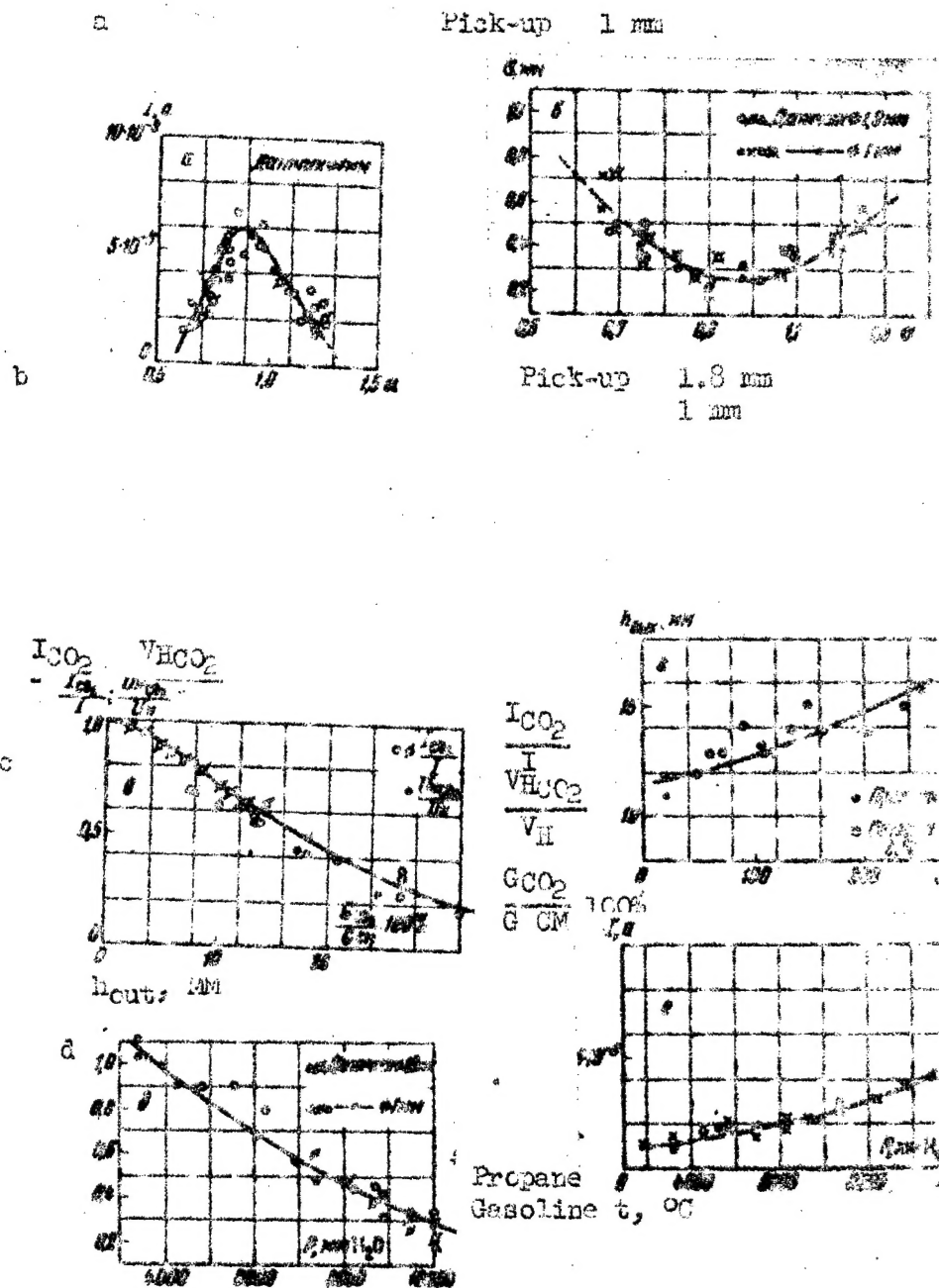


Figure 4. The dependence of the current and the width of the zone of unbalanced ionization in the laminar front of a flame upon the initial parameters of the mixture: a -- propane $T_1 = 20$ degrees C, $p = 1$ atmosphere, $U_0 = 13$ volts (pick-up with

e

Pick-up 1.8 mm
1 mm

P, mm H₂O

f

P, mm H₂O

diameter of 1 millimeter); b -- propane, $T_1 = 20$ degrees C, $\alpha = 0.9$ ($\delta \sim p^{-1.3}$); c -- propane; d -- propane and gasoline ($\delta \sim I \sim T_1^{0.53}$); e -- propane, $T_1 = 20$ degrees C, $\alpha = 0.9$ ($\delta \sim p^{-1.3}$); f -- propane, $T_1 = 20$ degrees C, $\alpha = 0.9$ (pick-up with diameter of 1 millimeter, $U_g = 13$ volts, $I \sim p^{1.3}$)

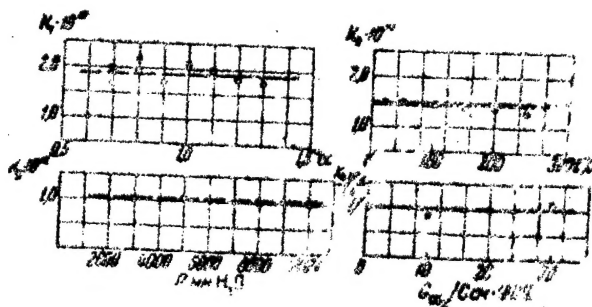


Figure 5

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